Scale Dependence of the Effective Matrix Diffusion Coefficient: Some Analytical Results

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Abstract

Matrix diffusion is an important process affecting solute transport in fractured rock, and the matrix diffusion coefficient is a key parameter for describing this process. Previous studies have indicated that the effective matrix-diffusion coefficient values, obtained from a number of field tracer tests, are enhanced in comparison with local values and may increase with test scale. In this communication, we develop analytical expressions for the effective matrix diffusion coefficient for two simple fracture-matrix systems, and demonstrate that heterogeneities in the rock matrix at different scales contribute to the scale dependence of the effective matrix diffusion coefficient.

1. Introduction

The matrix diffusion coefficient is an important parameter relating to transport in fractured rock, one that in many cases largely determines the overall solute transport behavior. Recently, several research groups have independently found that effective matrix diffusion coefficients much larger than laboratory measurements are needed to match field-scale tracer-test data (e.g., Neretnieks, 2002; Becker and Shapiro, 2000; Shapiro, 2001; Liu et al., 2004a). These observations imply that the effective matrix diffusion coefficient might be scale dependent and increase with test scale.

In addition to the observed enhancement, Liu et al. (2004b) and Zhou et al. (2005) examined many available in the literature and showed that scale dependence of the matrix diffusion coefficient was highly likely. This potential scale-dependence has important implications for large-scale solute transport in fractured rock. Liu et al. (2006) use numerical experiments to show that a combination of local flow loops (formed by locally connected small-scale fractures) and the associated matrix diffusion process, together with scaling properties in flow-path geometry, can cause the effective (or apparent) matrix diffusion coefficient to increase with travel distance, similar to the well known scale-dependence of dispersivity.

Given the complex nature of fracture/matrix systems, it is likely that more than flow path geometry alone contributes to the scale dependence. The major objective of this letter is to demonstrate that rock matrix heterogeneities may also contribute to the scale dependence of the effective matrix diffusion coefficient. In particular, we develop analytical expressions for the effective matrix diffusion coefficient for two idealized fracture-matrix systems. In addition to providing useful physical insights, analytical results can avoid curve fitting (often used to determine effective parameters from field tests), which involves uncertainties related to the non-uniqueness of parameter estimations using inverse modeling.

2. Effective matrix diffusion coefficient for a single fracture

Analytical solutions for solute transport (with matrix diffusion) in a homogeneous system involving a single fracture are commonly used to analyze and interpret field observations (e.g., Neretnieks, 2002). Here we determine the effective matrix diffusion coefficient for a single fracture involving heterogeneous rock matrix properties (Figure 1).

Our derivation is based on recent theories for determining solute transport in a single fracture with a wide range of retention processes (including matrix diffusion) and spatially variable flow and transport properties (Cvetkovic et al., 2004; Painter and Cvetkovic, 2005). According to these theories, the impulse-response function in the time domain for such a single fracture system with unlimited fracture spacing, which also may be viewed as the probability density distribution for a unit pulse input of conservative solute, is given by (Painter and Cvetkovic, 2005):

$$\gamma_l = \frac{H(t-\tau)B}{2\sqrt{\pi}(t-\tau)^{3/2}} \exp[\frac{-B^2}{4(t-\tau)}] \qquad [1]$$

where H is the Heaviside function, and t is time. The residence time τ is defined by

$$\tau = \int_0^l \frac{dl}{V} \qquad [2]$$

where l is the distance between the inlet and the location where a breakthrough curve is observed, and V is the water flow velocity along a fracture. Thus, l can be considered to represent the "scale" of the system. The parameter B is defined as

$$B = \int_0^l \frac{\phi\sqrt{D}}{b}\frac{dl}{V} \qquad [3]$$

where ϕ, D, and b are the matrix porosity, local matrix diffusion coefficient (molecular diffusion coefficient multiplied by tortuosity factor), and local half aperture, respectively. Note that the form of Eq. [3] is slightly different from the definition of parameter B given in Painter and Cvetkovic (2005) because of the difference between definitions of diffusion coefficients. The diffusion coefficient in Painter and Cvetkovic (2005) is the same as D multiplied by ϕ. The results based on Eqs [1] and [3] are consistent with the analytical solution for solute transport in a single fracture given by Neretnieks (2002). Approximating the tortuosity factor as equal to the matrix porosity (Liu et al., 2004b) allows D to be further expressed as

$$D = \phi D_0 \qquad [4]$$

where D_0 is the molecular diffusion coefficient in free water. Similarly, for a heterogeneous system, a representative local-scale matrix diffusion coefficient is given by

$$D_m = \phi_m D_0 \qquad [5]$$

where ϕ_m is the mean matrix porosity (corresponding to the geometric mean for a log-normal porosity distribution of local porosities).

For simplicity, we consider a fracture-matrix system (Fig. 1) with uniform matrix porosity values in the z direction, but spatially variable values in the x direction. Based on

Eq. [3], and keeping in mind that effective parameters are obtained by replacing the real, heterogeneous system with an effective, homogeneous system, we have:

$$B = l\phi_m \frac{\sqrt{D^*}}{bV} \qquad [6]$$

where D^* is the effective matrix diffusion coefficient. Combining Eq. [3] to [6] and considering bV to be constant yields

$$\sqrt{\frac{D^*}{D_m}} = \frac{1}{{\phi_m}^{1.5}} \frac{1}{l} \int_0^l \phi^{1.5} dl \qquad [7]$$

Consider ϕ to have a log-normal distribution with a standard deviation of σ_l (for the log of the matrix porosity) within the interval between x = 0 and l. Using the well-known expression for the arithmetic mean and moments of a log-normally distributed property, Eq. [7] can be further expressed as

$$\sqrt{\frac{D^*}{D_m}} = \exp(\frac{9}{8}{\sigma_l}^2) \qquad [8]$$

Obviously, if the log of rock matrix porosity is characterized by a stationary stochastic process, σ_l is not dependent on the scale l. However, a number of studies indicate that subsurface heterogeneity follows fractal-like long-range correlations that will give a scale-dependent σ_l (e.g., Molz et al., 1997).

While studies on porosity variability are relatively limited (Hassan et al., 1998), there are many studies in the literature on the permeability (K) variation and scaling of log(K). Therefore, it is of interest to relate the potential scale dependence of D^* to that for permeability. For the given rock matrix shown in Fig. 1, the effective matrix permeability (for water flow in the z direction) is the same as the arithmetic mean of permeability values between x =0 and l. Like many other researchers, we consider permeability as following log-normal distribution with a standard deviation σ_F for log(K). In this case, the effective permeability (K_e) is given as

$$K_e = K_g \exp(\frac{1}{2}\sigma_F{}^2) \qquad [9]$$

where K_g is the geometric mean of local permeability values.

Different relations between porous-medium permeability and porosity have been proposed in the literature. Such a relation was recently provided by Costa (2006):

$$K \propto \frac{\phi^m}{1-\phi} \qquad [10]$$

Considering that a rock matrix generally has small porosity values, the above equation can be further simplified as

$$K \propto \phi^m \qquad [11]$$

Combining Eqs. [8], [9] and [11] yields

$$\frac{D^*}{D_m} = \left[\frac{K_e}{K_g}\right]^{\frac{9}{2m^2}} \qquad [12]$$

For a typical value of $m = 3$ (Costa, 2006), the exponent in the Eq. [12] is equal to 0.5.

Eqs. [8] and [12] establish the intrinsic relations among effective matrix diffusion, effective matrix permeability, and porosity variability. It has been widely recognized that effective permeability is scale dependent and generally increases with test scale, and Eq. [12] indicates that the effective matrix diffusion coefficient should follow the same trend as the permeability.

3. Effective Matrix Diffusion Coefficient for Multiple Flow Channels

Whereas the focus of the previous section was on solute transport in a single fracture, flow and transport processes in fractured rock are in fact characterized by different flow channels within a fracture network (e.g., Neretnieks, 2002). Different flow channels may have different flow and transport properties. In this study, we use a simplified conceptual flow model to investigate the effects of interchannel heterogeneity of diffusive properties. Specifically, we consider a simplified multichannel system in which each flow channel has uniform properties and does not mix with any other channels except at influent and effluent points. These channels have the same length, width, fracture aperture, and other properties, but different matrix diffusion coefficients. In this case, the matrix porosity and matrix diffusion coefficient are not perfectly correlated.

We define $a = \frac{\phi_m \sqrt{D_m}}{b}$, and assume a to follow a normal distribution. When $t > \tau = \frac{x}{V}$ and for a pulse input (with mass M_0), the solute concentration for a channel is given by (Tang et al., 1981):

$$c(x,t) = \frac{M_0 \tau a}{bV\sqrt{\pi}(t-\tau)^{3/2}} \exp\left(-\frac{\tau^2 a^2}{t-\tau}\right) \qquad [13]$$

We assume the probability density function for a to be:

$$f(a) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{(a-\overline{a})^2}{2\sigma^2}\right] \qquad [14]$$

where σ is the standard deviation of a, and $\overline{a}$ is the mean of a. So the average concentration at the effluent point is (Zhang et al., 2006):

$$\overline{c}(x,t) = \frac{M_0 \tau \overline{a}}{bV\sqrt{\pi}(t-\tau+2\tau^2\sigma^2)^{3/2}} \exp\left[-\frac{\tau^2 \overline{a}^2}{t-\tau+2\tau^2\sigma^2}\right] \qquad [15]$$

The normal distribution for a is assumed here, simply because it allows for the derivation of an analytical solution for the average concentration. However, this analytical result is valid only for a small degree of heterogeneity, such that an insignificant portion of negative a values is included in Eq. [14]. To do so, we may impose the following limit to Eqs [14] and [15]:

$$\sigma' = \frac{\sigma}{\bar{a}} \leq \frac{1}{3} \qquad [16]$$

Eq. [15] can be further written as

$$\bar{c}(x,t) = \frac{M_0 TA}{bV\sqrt{\pi}(t-T)^{3/2}} \exp\left(-\frac{T^2A^2}{t-T}\right) \qquad [17]$$

where

$$T = \tau - 2\tau^2 \bar{a}^2 (\sigma')^2 \qquad [18]$$

$$A = \frac{\bar{a}}{1 - 2\tau\bar{a}^2(\sigma')^2} \qquad [19]$$

A comparison between Eqs. [13] and [17] indicates that Eq. [13] can be used to fit the average breakthrough curve (Eq. [17]) perfectly when T and A are treated as effective values for τ and a, respectively. (Note that Eqs. [18] and [19] are valid only when the denominator in [19] is positive.) In this case, the ratio of the effective matrix diffusion coefficient to a representative local-scale coefficient is

$$F_d = \left(\frac{A}{\bar{a}}\right)^2 = \left[\frac{1}{1 - 2\tau\bar{a}^2(\sigma')^2}\right]^2 \qquad [20]$$

For a given velocity, the residence time τ is proportional to the distance (or test scale). For typical values of V= 1m/d and $\bar{a}$ = 0.001 $s^{-0.5}$ and for σ'=1/3, Figure 2 shows F_d as a function of test scale. Again, this analytical result indicates the scale dependence of the effective matrix diffusion coefficient. It is also of interest to note that for a given test scale, an estimated effective matrix diffusion coefficient depends on velocity V. A large velocity would give a smaller residence time τ , and therefore a smaller effective matrix diffusion coefficient (see Eq. [19]).

The effective residence time T has a complex relation with the actual residence time τ . It initially increases and then decreases with τ . In their numerical studies, Zhang et al. (2006) were not able to detect the scale dependence of the effective matrix diffusion coefficient for a fracture system with interchannel heterogeneity, because they assumed T = τ . They also noted that their matches with the average breakthrough curves were not satisfactory and limited their simulations to those with relatively small heterogeneities.

4. Summary and Conclusions

In this letter, by developing analytical results for two fracture-matrix systems, we demonstrated that rock matrix heterogeneity is likely to contribute to the scale dependence of the effective matrix diffusion coefficient, with the effective coefficient increasing with scale. These analytical results can provide insightful relations between the effective matrix diffusion coefficient and other parameters such as porosity and hydraulic conductivity.

We derived relationships among the effective matrix diffusion coefficient, rock-matrix porosity variability, and effective rock-matrix permeability for a single fracture system. These relationships clearly indicate that the effective matrix diffusion coefficient and rock-matrix permeability follow a similar trend in terms of their relationship to test scale, while the permeability has been recognized in the literature to increase with test scale. We also developed an analytical expression for the effective matrix diffusion coefficient in a fracture-matrix system involving multiple flow channels, each of which has its own matrix diffusion coefficient. For a given water flow velocity, the expression again suggests that the effective matrix diffusion coefficient (resulting from the interchannel heterogeneity) keeps increasing with test scale. This solution also indicates that for a given test scale, a larger water velocity yieds a smaller effective matrix diffusion coefficient.

In natural systems, the scale-dependence of the effective matrix diffusion coefficient likely results from a combination of a number of different mechanisms. So far, we have identified two important mechanisms: the complexity of fracture network geometry that has been ignored in our current modeling practice (Liu et al. 2006), and the different-scale subsurface heterogeneity discussed in his letter. Both of these neglected mechanisms will be important in field-scale simulations of radionuclide and other solute transport in fracture/matrix systems.

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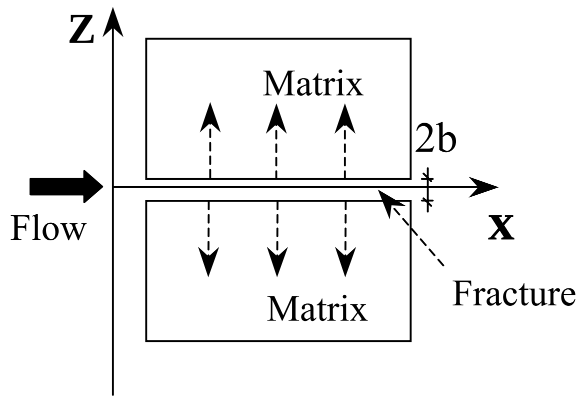

Fig. 1. Schematic of a single fracture system

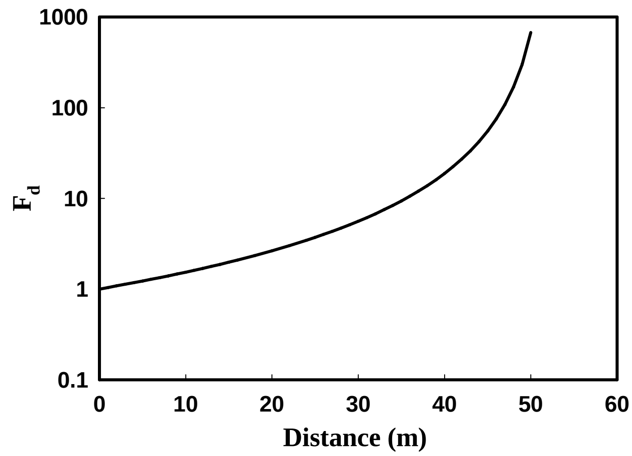

Fig. 2. Effective matrix diffusion coefficient as a function of travel distance for V= 1m/d, $\bar{a}$ = 0.001 $s^{-0.5}$ and for σ'=1/3